Review of “Multifactor colorimetric 1 analysis on pH-indicator papers: an optimized approach for direct determination of ambient aerosol pH”

This manuscript describes an improvement upon a recently published method (Craig et al. 2018) using image processing of colorimetric indicator paper to analyze the pH of atmosphere particles. The work is thorough and worthy of publication. There a few points I would suggest addressing and one major weakness to the manuscript. Overall, this method is an important step forward for determination of aerosol pH.

The largest concern with the manuscript is that it all of the analysis is with pipetted solutions and not with actual lab-generated or ambient aerosol as far as I can tell. Both Craig et al. 2018 and Coddens et al. 2019 from the Grassian laboratory looked at suspended aqueous aerosol that were then impacted onto colorimetric indicator paper. This led to some unique results (e.g. size dependence of pH), which make it not surprising that the 0.1 microlitre samples herein rapidly changed after pipetting on the paper. Even running just a few aerosolized samples to verify the selection of the specified pH paper would greatly strengthen the manuscript.

A minor is the justification of using 2 microlitres samples overall based on a high volume sampler pulling hundreds of lpm for a couple of hours. With that kind of flow rate and timing, a sample is unlikely to retain this amount of water due to drying and, at a minimum would be vastly altered at the end of sampling versus what was initially collected. Losses of semi-volatile inorganic (e.g. ammonium/ammonia) and organic (e.g. carboxylic acids like acetic acid) species would be expected in that sampling setup.

For Figure 2 it would be helpful to include both x- and y-error bars on the points, with x representing the uncertainty in the predicted pH and y the uncertainty in the pH probe/buffer measurements. This would help to know if the uncertainties include the regression line for the points that do not fall exactly on it.

This is a small point, but the term “outlier” is probably not the best for the point on Figure S4. If it is reproducible to the extent described it is by definition not an outlier. I think “anomalous” might be a better term, as this point would not be thrown out by the traditional Grubbs test of an outlier or other outlier tests.

The last point would be to that though the mention “anti-interference” it would be useful for the authors to see if their RGB method would work with brown carbon or black carbon samples (or some other chromophoric aerosol) that also contain secondary species and water.